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PCT/GB 41/05-100

09/857116



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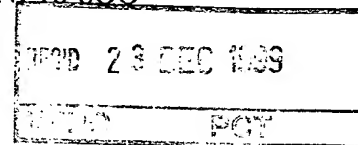
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Request for grant of a patent

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The Patent Office

Cardiff Road
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1. Your reference

JRL 1436 GB

2. Patent application number

(The Patent Office will fill in this part)

9826222.3

3. Full name, address and postcode of the or of each applicant (underline all surnames)

JOHNSON MATTHEY PUBLIC LIMITED COMPANY
2-4 COCKSPUR STREET
TRAFALGAR SQUARE
LONDON SW1Y 5BQ

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

GB

536262007

4. Title of the invention

IMPROVED REACTOR

5. Name of your agent (if you have one)

IAN CARMICHAEL WISHART

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

JOHNSON MATTHEY TECHNOLOGY CENTRE
BLOUNTS COURT
SONNING COMMON
READING RG4 9NH

Patents ADP number (if you know it)

7259312001

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and (if you know it) the or each application number

Country

Priority application number
(if you know it)

Date of filing
(day / month / year)

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

Date of filing
(day / month / year)

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? (Answer 'Yes' if:

a) any applicant named in part 3 is not an inventor, or

YES

b) there is an inventor who is not named as an applicant, or

c) any named applicant is a corporate body.

See note (d))

Patents Form 1/77

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Continuation sheets of this form

0

Description

4

Claim(s)

0

Abstract

0

Drawing(s)

1

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent (Patents Form 7/77)

Request for preliminary examination and search (Patents Form 9/77)

Request for substantive examination (Patents Form 10/77)

Any other documents (please specify)

11.

I/We request the grant of a patent on the basis of this application.

Signature

I C Wishart

Date 30 Nov 98

I C WISHART

12. Name and daytime telephone number of person to contact in the United Kingdom

MRS F E STRANGE 0118 924 2125

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IMPROVED REACTOR

The present invention concerns an improved reactor, more particularly it concerns a reactor particularly adapted for and suitable for certain selective oxidation reactions.

We have previously developed certain selective oxidation processes for the removal of residual carbon monoxide in hydrogen-containing gases intended as feedstock for fuel cells. Such processes are disclosed and claimed in for example, PCT/GB98/02873. It has become well established that hydrogen-fuelled fuel cells require very low proportions of carbon monoxide, which acts as a catalyst poison and degrades performance of the fuel cell. Desirably, the amount of CO is less than 10 ppm. If a liquid fuel is reformed to form a hydrogen-containing fuel, levels of CO are produced which may vary from about 0.5 to 5mol%, so that it becomes necessary to attenuate such CO levels. As mentioned in the above International Application, catalytic selective oxidation reactions are highly exothermic with the result that increasing reaction temperatures lead to increasing loss of valuable hydrogen due to competing reactions; temperatures lower than about 140 °C lead to a loss in selectivity, so that there is a rather narrow temperature "window" for highest conversions, which is desirably between 140 and 180 °C.

There remains a need for a catalytic selective oxidation reactor which permits good temperature control yet is simple, effective and inexpensive to manufacture. Although hydrogen "cleaned-up" by the reactor of the present invention may be used as fuel cell fuel, there are many other uses for such hydrogen. Also, although the reactor is particularly suitable for the cleaning up of reformat (which may be from methanol, other oxygenated fuels, gasoline or other hydrocarbon fuels), it is not limited to such use. Accordingly, the present invention provides a reactor suitable for catalytic selective oxidation, comprising a reactor vessel with counter current cooling and at least one stage, each such stage provided with an inlet for a first feedstock, e.g. CO contaminated hydrogen, an inlet for a second feedstock, e.g. oxygen-containing gas, gas mixing means and a catalytic reaction zone, preferably comprising a


selective oxidation catalyst deposited on a substrate having good heat conduction properties and desirably having very low pressure drop.

The invention further provides a process of catalytic selective oxidation, comprising the use of a reactor as defined above.

The reactor, in its most preferred embodiment, has four stages and this is effective to reduce a reformat having a CO content of 1 to 2% to below 10ppm. In other embodiments, there may be 2 to 10 stages.

The reactor is suitably in the form of a vessel in which the reaction zones are generally annular in shape, and provided with internal and/or external liquid cooling. cooling is preferably provided by internal cooling using water, which is suitably under pressure to prevent boiling at the temperatures generated. Other liquid coolants, or forced air cooling may be used if desired. According to specific reactor designs, heat removal may be enhanced by increasing heat exchange surface areas by corrugation or other established means. The rate of heat removal may be altered by altering the flow of coolant and/or altering the quantity of heat removed from the coolant by means of a heat exchanger.

Each reaction zone suitably comprises a metallic catalyst support ("monolith") of generally honeycomb construction, as is in common use in automobile exhaust gas catalysis. These may have 50 to 1200 cells per square in cell density, preferably 200 to 600 cpsi, and may be of a variety of types of stainless steel or other metals. In general, the present invention does not result in the reaction stages reaching the higher temperatures generally met with in exhaust gas catalysis (e.g. 500-900°C), but the material should be capable of resisting temperature excursions. An alternative reaction zone design utilises what has become known as "static mixers" which combine a very high degree of gas agitation and mixing, resulting in non-laminar flow, with relatively low pressure drop. Such static mixers may be manufactured from a variety of metals and are commercially available. However, it is presently preferred to use a



honeycomb monolith. It is to be noted that honeycomb substrates do not provide significant gas mixing, unless modified.

The reaction zone desirably carries a selective oxidation catalyst comprising a platinum group metal, especially comprising platinum and/or rhodium, carried on a high surface area metal oxide support, such as alumina. The metallic catalyst support may be coated by generally known methods with a slurry of oxide support, dried and fired, and then impregnated with the catalytically active components, before a final firing. Such procedures are in themselves known, and do not form part of this invention.

The invention will now be described with reference to the accompanying drawing in which Fig 1 shows a schematic axial cross-section of one embodiment of the present invention.

An essentially cylindrical metal vessel, 1, made by pressing two halves and seam-welding, has a main gas inlet, 2, for reformat gas, and an outlet, 3, for cleaned gas. The vessel has a number of annular reaction zones, 4, each consisting of an annular wound corrugated catalyst support. The vessel further has an internal water jacket, 5, between a spacer, 5a, and the shell, and this is supplied with cooling water by means of a water circuit comprising a circulating pump, 6, and a heat exchanger, 7. The heat exchanger is desirably used to recover heat in an integrated system e.g. in an integrated combined heat and power system. The vessel further is provided with four air inlets, 8, each providing pre-determined (for a steady-state operation) levels of air (that is reaction oxygen; air is generally suitable). Although not preferred, controlled amounts of oxygen, according to one or more feedback or feed forward controls detecting gas flow levels and CO levels may be used. Downstream of each air inlet is a gas mixing zone, 9a, 9b, 9c and 9d. Zone 9a is suitably a static mixer, but zones 9b, 9c and 9d are preferably alternating annular mixing vanes, 10, and discs, 11, having gas flow holes, as illustrated in Figs 2 and 3. It should be noted that for simplicity only four vanes and four holes are shown, but more, e.g. six or eight, may be used. Such vanes and discs may be manufactured by pressing to form a sleeve which may be fitted and brazed or welded onto the jacket 5 with

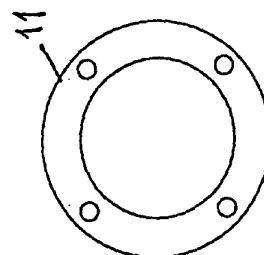
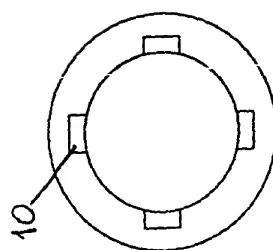
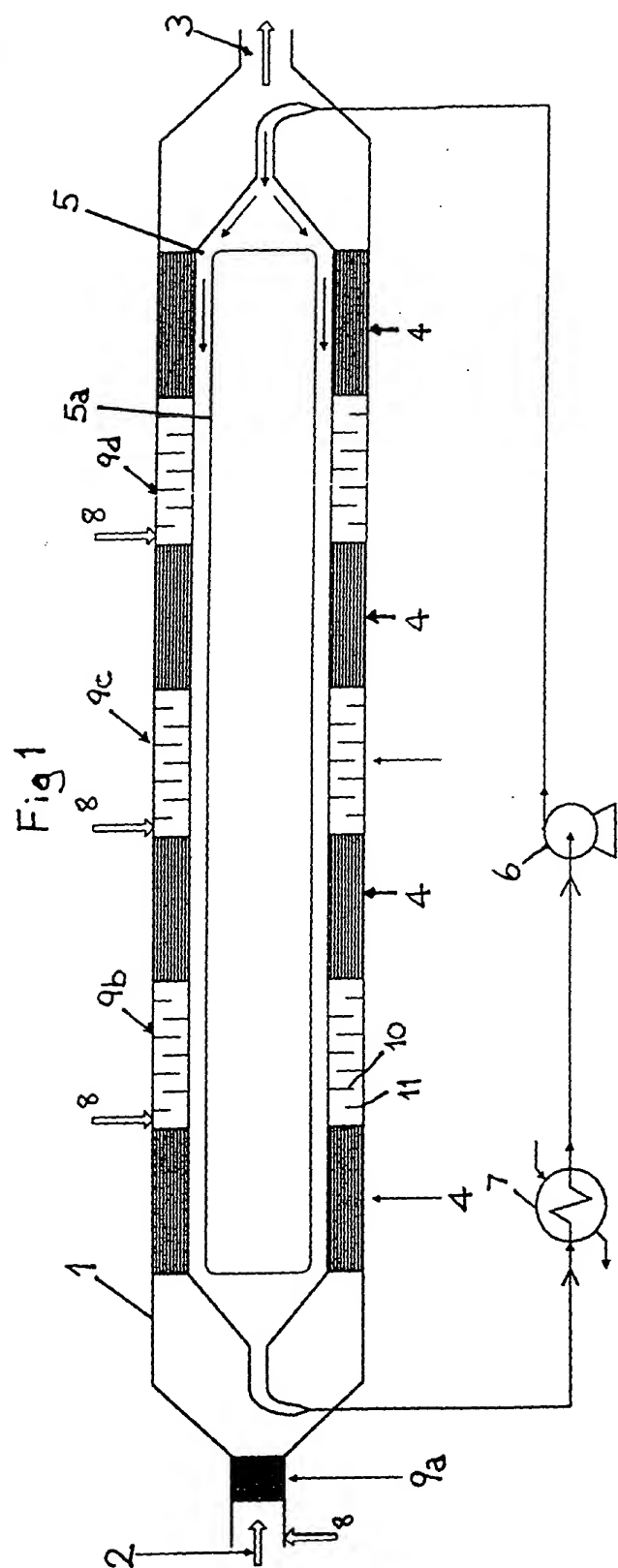
the intention that each gas mixing zone also serves to remove heat from the reaction zone, leaving each reaction zone. Although the schematic drawing of the reactor shows each reaction zone and gas mixing zone to be of the same size, these may be designed to differ according to the amount of heat to be removed. Additionally, each reaction zone may incorporate different and/or different loadings, of catalyst.

Each metal catalyst support monolith suitably has 300–400 cells per sq.in. and is coated with a washcoat comprising ~36wt% alumina solids of D50 of approx $5\mu\text{m}$ and D90 of approx $16\mu\text{m}$, to deposit approx 3g/in^3 . After drying and firing, the coated monolith was impregnated with a platinum salt solution to result in 5% by wt of Pt deposited on the alumina. The resulting coated monolith was again dried and fired before being assembled into the reactor.

Initial studies indicate that a reformat containing 1-2% CO in admixture with H_2 , CO_2 , N_2 , water vapour with a small proportion of unreacted reformer feedstock (e.g. CH_3OH , CH_4 , gasoline), at 180°C may be converted to a feed gas suitable for a fuel cell, containing below 10ppm CO, and at 140°C .

The reactor of the invention is compact and exhibits very low pressure drop. Manufacturing costs are relatively low. The reactor is believed to be suitable for mobile or automobile uses as well as stationary uses. It is presently envisaged that the reactor shell will be lagged to control heat loss and prevent injury. The reactor can be manufactured simply and inexpensively, using pressed and welded parts. It is desirably operated to achieve low pressure drop, and the particular embodiment illustrated is designed for a gas hourly space velocity of approx. $30,000\text{h}^{-1}$ gas throughput.

The reactor as described may be altered in a variety of ways without departing from the central inventive concept.



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Johnson Matthey plc

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